Dynamics of Random Spin Systems

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ABSTRACT

We present inelastic neutron scattering experiments on three prototypical random magnets. For the dilute, insulating antiferromagnet $\text{Rb}_2\text{Co}_x\text{Mg}_{1-x}\text{F}_4$, the randomness has purely geometrical consequences, and the anomalous dynamical behavior which we observe for x close to the magnetic percolation threshold is due to the fractal nature of percolating networks. Comparison of inelastic and quasielastic scattering data demonstrate the dynamical nature of the spin glass transition in amorphous, metallic MnSi . Finally, we show the coexistence of spin waves and static spin fluctuations near the crossover from ferromagnetic to spin glass behaviors in amorphous $(\text{Fe}_x\text{Mn}_{1-x})_{75}\text{P}_{10}\text{B}_6\text{Al}_3$.

Keywords: Percolation, Spin Glass, Spin Waves, Central Peak, Disorder

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I. INTRODUCTION

While the static properties of random systems are quite interesting, many of the qualitative effects unique to random systems are dynamical. In this paper, we show that inelastic neutron scattering is a powerful probe of such effects. Specifically, we describe experiments on three prototypical random magnets. The first and simplest is $Rb_2Co_2Mg_{1-x}F_4^{(1)}$, where the randomness does not enhance competing interactions and thereby induce frustration; thus, the ground state for any x is easily specified and the static properties⁽²⁾ are well understood. Nonetheless, the disorder has profound geometrical consequences which produce dynamical effects not observed for the corresponding pure system.⁽³⁾ The second system we consider is an archetypical, *concentrated* metallic spin glass, amorphous $MnSi^{(4)}$, where the interesting dynamical effects are due not to geometry, but to competing interactions. Finally, we consider a system, $(Fe_{1-x}Mn_x)_{75}P_{10}B_5Al_5$, where both geometry and frustration play important roles.⁽⁵⁾

The experiments described below were all performed using triple axis spectrometers at the Brookhaven High Flux Beam Reactor (HFBR). Monochromators and analyzers are vertically bent pyrolytic graphite (PG). Because relatively high resolution (< 0.15 meV Full Width at Half Maximum [FWHM]) was required, neutron energies were below 5 meV; Be filters were used to eliminate higher-order contamination. For the instruments located at thermal beam ports, spectra were collected by varying the final energy E_f for fixed incident energy $E_i \geq 3.5$ meV. One spectrometer is installed at a cold neutron source where, E_i can be set to energies as low as 2.3 meV. For $E_f = 2.5$ meV and the configuration 30'-PG1-40'-PG2-30'-SAMPLE-40'-PG3-20' (the instrument is equipped with a double monochromator), the resolution is $37 \mu eV$ (FWHM). As will become apparent below, further examination of random spin systems will generally require the better resolution of the back-scattering and neutron spin echo techniques.

II. $\text{Rb}_2\text{Co}_c\text{Mg}_{1-c}\text{F}_4$

Pure Rb_2CoF_4 is one of the best realizations of the two-dimensional Ising model, with a Néel point of 103K and critical exponents as predicted by Onsager.⁽³⁾ The dominant interactions couple nearest neighbor Co^{2+} ions, which are arranged on square lattices within the basal planes of this tetragonal compound. Random substitution of non-magnetic Mg^{2+} ions for the Co^{2+} ions should therefore lead to behavior characteristic of magnetic percolation in two dimensions. To check this, Cowley and co-workers carried out a thorough study of the instantaneous spin correlations in the dilution series $\text{Rb}_2\text{Co}_c\text{Mg}_{1-c}\text{F}_4$.⁽²⁾ They found that long-range antiferromagnetic order does indeed disappear as c is scanned through the nearest-neighbor percolation threshold ($c=c_p = 0.593$) for a square lattice. Furthermore, percolation theory gives an excellent account of the magnetic correlations observed.⁽²⁾

While the static behavior near the magnetic percolation threshold has been surveyed for a variety of two and three-dimensional systems and is by now well-understood, the corresponding dynamical behavior, particularly at low frequencies, has received almost no theoretical or experimental attention. We consequently set out to measure the dynamical spin correlation function in $\text{Rb}_2\text{Co}_c\text{Mg}_{1-c}\text{F}_4$ for $c=0.58 \approx c_p$. We selected this compound for study not only because it is a dilute two-dimensional Ising antiferromagnet and thus easily simulated by computer, but also because the critical dynamics of pure Rb_2CoF_4 had been established previously.⁽³⁾

Fig. 1 shows data obtained at the cold source spectrometer. The scattering vector was fixed at $Q=(1, 0, 0.4)$, which lies on the line in reciprocal space where the Bragg condition for two-dimensional antiferromagnetic order is satisfied. Thus, the experiment measures the Fourier transform of the auto-correlation function $[M(t) M(t + \tau)]$ for the staggered magnetization $M(t)$. Lorentzian profiles $A\Gamma/(\Gamma^2+(\hbar\omega)^2)$, corrected for background and resolution effects, are in excellent agreement with the data, as can be seen in Fig. 1. Therefore, to within experimental uncertainty, $[M(t) M(t + \tau)]$ is a simple exponential characterized by the decay rate Γ . Fig. 2 shows the reduced decay rate Γ/kT plotted as a function of the inverse

magnetic correlation length κa . The power law $\Gamma/kT \sim (ka)^z$ with $z = 2.4 \pm 0.1$ fits the data quite well. Given the limited temperature range over which we could measure Γ/kT reliably, the important result is not the precise value of z , but the fact that it is much higher than that (1.7) measured for pure $\text{Rb}_2\text{Co F}_4^{(9)}$. Furthermore, the conventional theory of the kinetic Ising model⁽⁶⁾, which yields the experimentally observed z for pure $\text{Rb}_2\text{Co F}_4^{(9)}$, gives $z = 2\eta \approx 1.67$ for two-dimensional magnetic percolation.⁽²⁾

The $T=0$ transition for a percolating magnet is similar to phase transitions in pure magnets in that its essential feature is the divergence of a single correlation length. The dynamical behavior, however, can be expected to be somewhat unconventional because the travel times τ for disturbances in fractal systems as ramified as percolating networks rise more rapidly with (Euclidean) distance l than they do in ordinary Euclidean spaces.⁽⁷⁾ An anomalous rise of τ with l would indeed account for the large value of z which we measure for $\text{Rb}_2\text{Co}_x\text{Mg}_{1-x}\text{F}_4$. Recent theoretical work⁽⁸⁾ suggests that if the measurements were extended to lower temperatures, the exponent z would be found to be temperature dependent, diverging as T approaches zero. Back-scattering experiments to test this dramatic prediction are currently in progress.⁽⁹⁾

III. Amorphous MnSi

Crystalline $\text{MnSi}^{(10)}$ is a strongly fluctuating (even as $T \rightarrow 0$) itinerant magnet which undergoes a transition to a long period (180 Å) helimagnetic state at $T_A = 29.5\text{K}$. In amorphous MnSi , the distance between the Mn atoms are random, leading to interactions between the spins which are random in sign. The result is that a-MnSi is a concentrated metallic spin glass⁽¹¹⁾ whose bulk properties are remarkably similar to those of the classical, dilute RKKY system, CuMn .

Fig. 3 shows the neutron scattering intensity for various spectrometer configurations. Frame (a) represents data taken with a double axis spectrometer, which probes the instantaneous spin correlation function $S(Q)$. While there is a gradual rise in signal as the

temperature is reduced, there is no visible anomaly near the spin glass temperature $T_g = 24\text{K}$, determined from a.c. susceptibility measurements.⁽¹¹⁾ The solid line represents $kT\chi_{dc}$ (χ_{dc} is the measured, bulk dc susceptibility), which for classical paramagnets is directly proportional to $S(Q=0)$. For $T > 40\text{K}$, the susceptibility and neutron data are in excellent agreement, while for $T < 40\text{K}$, $kT\chi_{dc}$ falls below the neutron data. Behavior of this type is expected if there is a spin-glass transition below which $S(Q,\omega)$ contains an elastic term $k(\omega)q_0$, due to spin freezing,¹² in addition to the contribution proportional to χ_{dc} , due to spin relaxation. The quasielastic intensity measured near the forward direction must then be proportional to the sum $kT\chi_{dc} + q_0$. In principle, q_0 is the elastic signal observed with a triple axis spectrometer, while χ_{dc} can be derived from the inelastic spectrum. Figures 3(c) and (d) show the elastic scattering measured for resolutions of 150 and 10 μeV (FWHM), respectively. Gratifyingly, our highest resolution elastic scattering data (Fig. 3(d)) do correspond to roughly the difference between the quasielastic scattering and $kT\chi_{dc}$ (Fig. 3(a)). Fig. 3(b) shows the inelastic scattering observed at $\hbar\omega = 0.25\text{ meV}$; these data provide a measure of the a.c. response at $6 \times 10^7\text{ kHz}$. As the relaxation frequency for the spin glass crosses 0.25 meV (at $T \approx 50\text{K}$), the response reaches a maximum.

The neutron scattering data for a-MnSi clearly demonstrate the dynamical nature of the SG transition: the instantaneous spin correlations $[S_i(t)S_j(t)]$ evolve negligibly with T ; instead, the essential quantity is the local auto-correlation function $[S_i(t)S_i(t+\tau)]$, which approaches q_0 as $\tau \rightarrow \infty$. Thus, spin glasses are very different from percolating magnets, where, as for conventional magnets, both the instantaneous spin correlations and the local auto-correlations undergo non-trivial and related (via dynamical scaling⁽⁶⁾ or its new generalizations⁽⁸⁾) evolutions with temperature.

IV. $(\text{Fe}_{1-x}\text{Mn}_x)_{75}\text{P}_{10}\text{B}_6\text{Al}_3$

For $0.15 < x < 0.35$, $(\text{Fe}_{1-x}\text{Mn}_x)_{75}\text{P}_{10}\text{B}_6\text{Al}_3$ displays paramagnetic (PM), ferromagnetic (FM), and then spin glass (SG) behavior as the temperature is reduced.⁽¹³⁾ In contrast to what is observed for a-MnSi, $S(Q)$ is strongly dependent on Q ⁽¹⁴⁾, and, as a function

of T , undergoes a dramatic evolution (See Fig. 4) mirroring that of the alloy's bulk susceptibility.⁽¹³⁾ Fig. 5 shows inelastic scattering data for the $x=0.2$ alloy, which exhibits a Curie point at $T_c = 340\text{K}$ and an FM-SG cross-over temperature of $T_f = 34\text{K}$.^(13,14) At high temperatures (frames (a) and (b)), well-resolved spin wave peaks dominate the spectra. As T is lowered, the peaks move first to higher energies, as they should for an ordinary FM. However, for $T < 150\text{K}$, the spin wave energies decrease with T . Furthermore, for $T < 50\text{K}$, a strong peak appears for $\hbar\omega = 0$ meV. While true inelastic scattering persists to the lowest T (5K), the integrated quasielastic intensity becomes significantly larger than that of the inelastic scattering as T is reduced below 30K. The central peak is resolution-limited for the highest resolution (26 μeV FWHM) used. It is also very Q -dependent, decaying as $Q^{-\alpha}$ with $\alpha > 2$.⁽⁶⁾ Thus, our experiment demonstrates the coexistence of dynamical fluctuations describable as heavily damped spin waves and long-range fluctuations with relaxation times exceeding 10^{-9} sec. It is worth noting that similar results have been obtained for other alloys exhibiting FM-SG transitions.⁽¹⁵⁾

It is apparent that $(\text{Fe}_{1-x}\text{Mn}_x)_{75}\text{P}_{10}\text{B}_5\text{Al}_3$ is considerably more complicated than $\alpha\text{-MnSi}$ and $\text{Rb}_2\text{Co}_{1-x}\text{Mg}_x\text{F}_4$. However, our understanding^(3,14) of $(\text{Fe}_{1-x}\text{Mn}_x)_{75}\text{P}_{10}\text{B}_5\text{Al}_3$ relies on a combination of concepts borrowed from discussions of the percolation and spin glass problems. In particular, the random alloy is subdivided into connected ferromagnetic and spin glass networks, with characteristic temperatures T_c and T_f . As long as $T_c > T \gg T_f$, the system will behave much as a diluted FM, with both the magnetization M and spin wave stiffness D_{eff} rising with decreasing T . As T approaches T_f , the relaxation spectrum of the SG network will overlap the FM spectrum. The two networks will therefore interact strongly to yield the observed decrease in M and D_{eff} . Finally, at low T , the "frozen" SG network applies a static random field to the FM network; random fields⁽¹⁶⁾ are known to produce strong quasielastic scattering of the type⁽¹⁴⁾ seen in $(\text{Fe}_{1-x}\text{Mn}_x)_{75}\text{P}_{10}\text{B}_5\text{Al}_3$.

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Figure Captions

Fig. 1 Constant-Q inelastic spectra, all collected for collimations and neutron energy indicated in (a)

The dashed horizontal lines represent the constant background, the solid lines correspond to the profile computed, as described in text, using the indicated values of Γ , and the dashed curve in (a) corresponds to the same profile (2) calculated without the elastic background term.

From Ref. 1

Fig. 2 Normalized relaxation rate Γ / kT plotted against reduced inverse correlation length κa . At the dashed curves, the FWHM of the resolution function exceeds Γ by a factor of 4. The open and filled circles correspond to the two different spectrometers described in text.

From Ref. 1

Fig. 3 Intensity of scattered neutrons measured with use of (a) double-axis and triple-axis (b-d) spectrometers. Counting times were 12, 180, 50, and 60 min. respectively. The left-hand vertical scales correspond to the number of magnetic and nonmagnetic scattering events actually observed. The zeros of the right-hand scales represent the nonmagnetic-background levels. For (a) [(b)-(d)], unity corresponds to the net [elastic,

$E_i = E_f$ scattering observed with the given spectrometer at the lowest temperature. Thus, using the right-hand scale, the intensities in (b) and (c) can be compared directly.

From Ref. 4

Fig. 4 Temperature dependence of SANS intensity measured at various momentum transfers q for $(Fe_{1-x}Mn_x)_{75}P_{15}B_0Al_3$.

From Ref. 14

Fig. 5 Evolution with temperature of inelastic spectra for $Q=0.06 \text{ \AA}^{-1}$ and $x=0.20$. Corrections have been made for a flat (Q - and E - independent) background. 100 monitor counters correspond to counting times of roughly 30 sec. for (a), (c), and (d), and 1 min. for (b).

From Ref. 5

FIGURE 1

FIGURE 2

FIGURE 3

FIGURE 4

FIGURE 5